CHARACTERIZATION OF A PULSED X-RAY SOURCE FOR FLUORESCENT LIFETIME MEASUREMENTS*

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ABSTRACT

To search for new, fast, inorganic scintillators, we have developed a bench-top pulsed x-ray source for determining fluorescent lifetimes and wavelengths of compounds in crystal or powdered form. This source uses a light-excited x-ray tube which produces x-rays when light from a laser diode strikes its photocathode. The x-ray tube has a tungsten anode, a beryllium exit window, a 30 kV maximum tube bias, and a 50 μA maximum average cathode current. The laser produces 3×10^{7} photons at 650 nm per ~100 ps pulse, with up to 10^{7} pulses/sec. The time spread for the laser diode, x-ray tube, and a microchannel plate photomultiplier tube is less than 120 ps fwhm. The mean x-ray energy at tube biases of 20, 25, and 30 kV is 9.4, 10.3, and 11.1 keV, respectively. We measured 140, 230, and 330 x-ray photons per laser diode pulse per steradian, at tube biases of 20, 25, and 30 kV, respectively. Background x-rays due to dark current occur at a rate of 1×10^{6} and 3×10^{6} photons/sec/steradian at biases of 25 and 30 kV, respectively. Data characterizing the x-ray output with an aluminum filter in the x-ray beam are also presented.

I. INTRODUCTION AND MOTIVATION

This pulsed x-ray source was developed to aid an ongoing search for new scintillators for gamma ray detection. Faster and brighter scintillators would improve the scintillation detector recovery time and energy resolution, and could also reduce the cost. Our previous efforts to find new scintillators used an electron synchrotron in single-bunch mode to measure the x-ray excited fluorescence of over 400 compounds [1, 2], but use of a synchrotron is costly and access limited. This bench-top pulsed x-ray source reduces the cost and increases the ease of making these measurements.

The design priorities for the pulsed x-ray source included compact size, low cost, the capability to observe fluorescence from powders as well as crystals, and the capability to determine scintillation time structure on the order of tens of picoseconds. The incorporation of a light-excited x-ray tube manufactured to produce very brief (≈ 100 ps) pulses of x-rays [3] made these priorities realistic. The design of the pulsed x-ray source is described in [4]; the primary purpose of this paper is to characterize its performance.

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II. SYSTEM DESIGN

A. General Design

The two primary components of the pulsed x-ray source are the light-excited x-ray tube and the diode laser, shown diagramatically in Figure 1. The x-ray tube is essentially a single-stage photomultiplier tube, with a photocathode which releases electrons when light is absorbed. The electrons are accelerated across 30 kV (typically) into a tungsten anode, where bremsstrahlung x-rays are produced upon impact.

The light-excited design of the x-ray tube makes it possible to generate short pulses of x-rays simply by directing short pulses of light onto the photocathode of the tube. The repetition rate of the x-ray pulses can be varied by changing the repetition rate of the light pulses. For the pulsed x-ray source presented here, a diode laser was used as the light source because of its short laser pulse duration, easily varied repetition rate, and relatively low cost. X-ray pulses with arbitrary time structure can be created by modulating the incident light source to have the desired structure. The incident light is easily modulated (while x-rays are quite difficult to modulate)

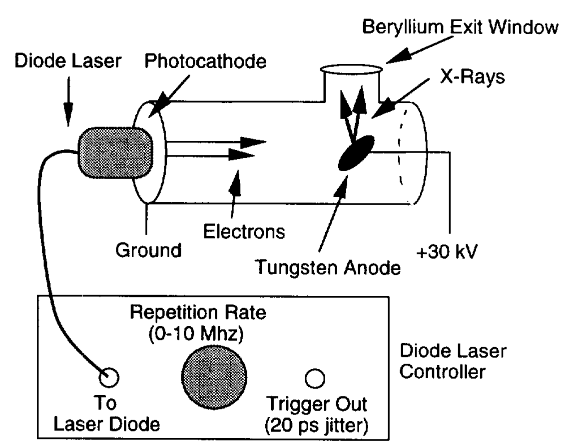

Figure 1. Schematic of the pulsed x-ray source. A laser diode excites the photocathode with <97 ps pulses at ≤10 MHz.

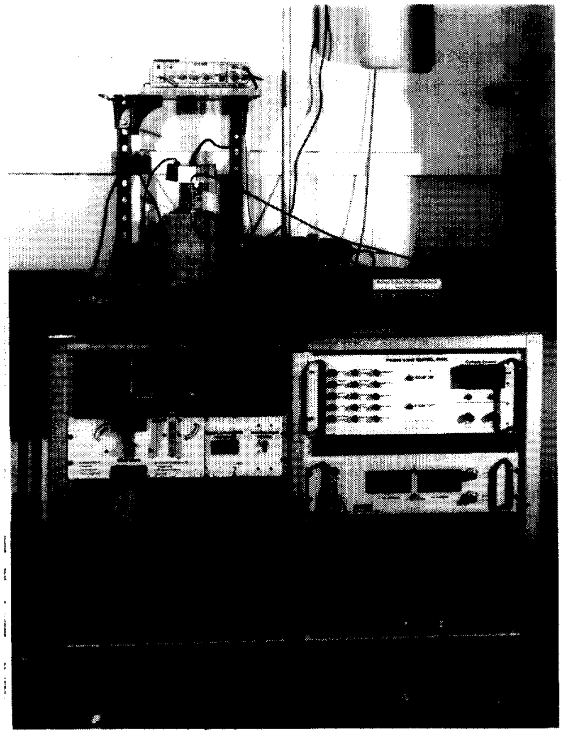

Figure 2. Photograph of the pulsed x-ray source.

and the estimated broadening due to the x-ray tube is only 31 ps. Other potential applications for x-ray sources with controlled time structure include characterizing afterglow for x-ray CT scanners or freezing motion (such as a beating heart or a moving part) in x-ray imaging.

Figure 2 is a photograph of the pulsed x-ray source. The steel box on the table top is the sample chamber, and directly behind it is the x-ray tube. The laser diode is mounted above the x-ray tube, and the laser diode controller is on the stand above the table top. The sample chamber is evacuated by the pump under the table top. As the system is configured in this photograph, scintillation light passes through the quartz telescope on the right side of the sample chamber to a microchannel plate photomultiplier tube. The x-ray tube high-voltage power supply and the control panel, which includes extensive safety interlocks and a current meter for monitoring the x-ray tube cathode current, are both mounted in the lower right portion of the table.

Table I. Characteristics of Hamamatsu PLP-01 Light Pulser with C3551-01 Controller and LDH065 Laser Diode Head

Emission wavelength	650 nm
Peak power	>100 mW
Average power (max)	0.1 mW
Pulse width	<97 ps fwhm
Pulse repetition rate	dc to 10 MHz
Photons per pulse	> 10^7
Timing pulse jitter	±10 ps

B. Component Characteristics

The characteristics of the diode laser and light excited x-ray tube are summarized in Table I and Table II.

III. SYSTEM CHARACTERIZATION

A. System Impulse Response

We expect the width of the x-ray pulse to be 84 ps fwhm (full-width at half-maximum), based on the 78 ps fwhm width of the diode laser pulse convolved with the theoretically estimated (based on the tube design) time spread of the x-ray tube of 31 ps fwhm. The fluorescent photon detection apparatus (a microchannel plate photomultiplier tube or MCP–PMT) has a 41 ps fwhm single photon impulse response, determined by exciting the MCP–PMT with a 30 ps fwhm laser pulse, then subtracting (in quadrature) the laser contribution to the measured 51 ps fwhm total width. A consistency check is obtained by exciting the MCP–PMT with the diode laser. The predicted value of the response of 84 ps agrees with the measured 97 ps fwhm time spread. Reference [5] further discusses the predicted impulse response.

We are not equipped to directly measure the x-ray pulse width. To approximate this measurement, we added an ultra-fast scintillator and a MCP–PMT to the system in order to obtain fluorescence data as an upper limit on the impulse response of the laser diode, the x-ray tube, and the photomultiplier tube. The scintillator used was malachite green oxalate in crystal form, which likely has a fluorescence decay time of 10–30 ps (based on fluorescence decay times of malachite green solutions [6]). The observed fluorescence time structure (collected for 235,000 seconds) is shown in Figure 3, and has a width of 120 ps fwhm. The peak at 400 ps is due to after-pulsing in the diode laser — we are working with the manufacturer to eliminate this. We conclude that the system impulse response for the laser diode, x-ray tube, and MCP–PMT is less than 120 ps fwhm, which is consistent with the above estimate of 84 ps fwhm for the x-ray pulse width.

Table II. Characteristics of Hamamatsu N5084 Light-Excited X-Ray Tube

Overall tube length	152 mm
Tube Diameter	52 mm
Photocathode	S-20
Quantum efficiency @ 650 nm	> 10%
Photocathode diameter	12 mm
Target material (anode)	Tungsten (45°)
Output window material	Beryllium
Output window diameter	20 mm
Output window thickness	0.5 mm
Cooling	Natural air
Tube voltage (max)	30 kV
Average tube current (max)	50 μA
Peak tube current	2 A (2 μs duration)
Photocathode photon rate (max)	3×10^{15}/s @ 10% QE

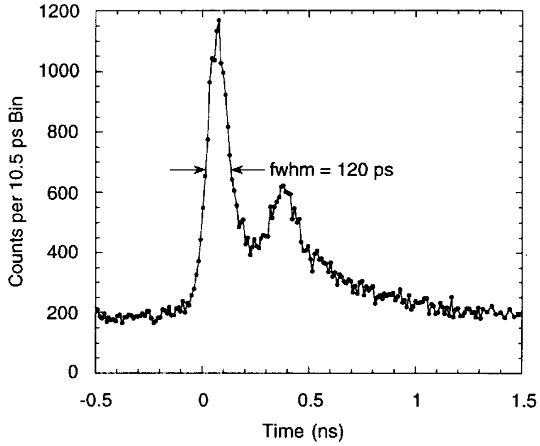

Figure 3. Observed fluorescence time spectrum for malachite green oxalate crystals, which provides an upper limit of 120 ps for the system impulse response.

B. Energy Spectra of X-Rays

The measured spectral output of the x-ray source was obtained using a lithium-drifted silicon detector. The detector was cooled to liquid nitrogen temperature and had an energy resolution of 180 eV fwhm at 2 μs peaking time. The detector was positioned 14 cm from the x-ray tube anode, with a lead pinhole collimator placed over the detector to limit the count rate. Each spectrum was collected over 1800 seconds using a 200 kHz laser diode pulse repetition rate. Corrections were made for the energy dependent x-ray absorption probability in the 6 mm thick detector (a 21% adjustment at 30 keV). The data in these energy spectra were used for the remaining figures. The background flux of the x-ray source in the absence of laser light has been subtracted from the following figures, with an extrapolated value used for the case with a 32 kV tube bias and no filtration. This background flux will be discussed in a later section.

Figure 4 shows the spectral output of the pulsed x-ray

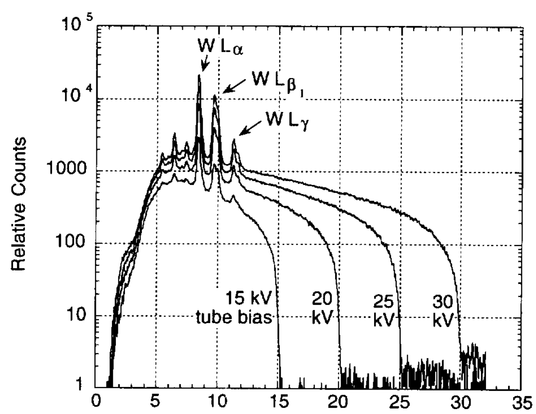

Figure 4. Energy spectra of the x-ray source vs. tube bias.

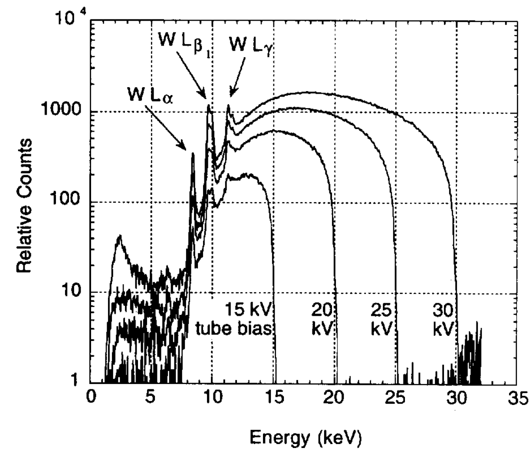

Figure 5. Energy spectra of the x-ray source with a 0.51 mm aluminum filter.

source at various tube biases. These spectra are typical for x-rays from a tungsten anode x-ray tube, with bremsstrahlung radiation at higher energies and characteristic tungsten peaks around 10 keV. The additional peaks between 5 keV and 8 keV correspond to iron, chromium, manganese, and copper characteristic peaks from fluorescence of various system components.

In practice, we place a 0.51 mm aluminum filter in the x-ray beam to absorb low-energy x-rays. These x-rays would be absorbed in the quartz cuvette holding the sample, but would not contribute to the fluorescence signal from the sample and would generate a low level of background fluorescence from the quartz. The measured spectra after filtering are shown in Figure 5.

Figure 6 shows the mean x-ray photon energies as a function of tube bias for the spectra shown in Figures 4 and 5 and for spectra obtained at a tube bias of 32 kV. As expected, the aluminum filter significantly increases the mean photon energy.

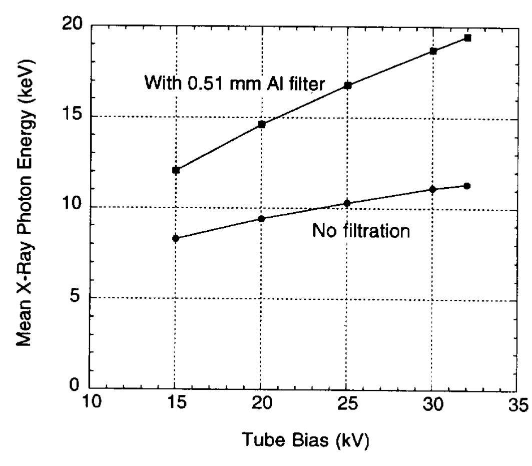

Figure 6. Mean energy of x-ray photons at various tube biases, with and without the aluminum filter.

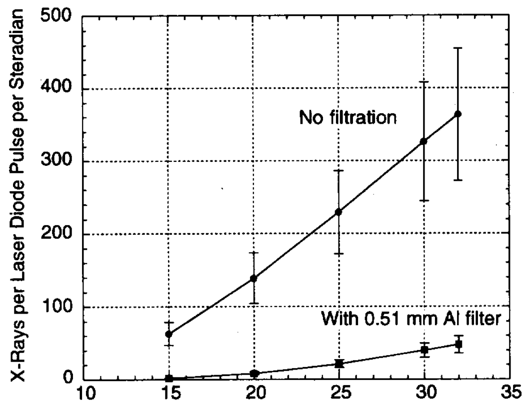

Figure 7. X-ray flux resulting from laser pulses. The 25% error bars (in this figure and figures 8 and 9) are based on the estimated precision of the lead collimator pinhole diameters.

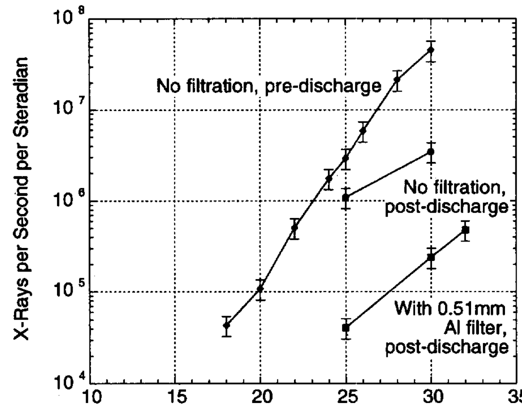

Figure 9. Background flux in the absence of laser light.

C. X-Ray Flux

The relative flux of x-ray photons was determined by integrating the counts in the spectra and making minor corrections (<2%) for system dead time. This relative flux was converted to absolute flux (x-rays per laser pulse per steradian) using the distance from the x-ray tube anode to the detector, the size of the lead collimator pinhole, and the laser pulse repetition rate. The flux, shown in Figure 7, increases nearly linearly with tube bias.

Multiplying the flux of Figure 7 by the mean photon energy of Figure 6 yields the energy deposited per laser pulse per steradian, shown in Figure 8. The higher mean photon energy for the filtered case partially compensates for the lower flux, so that the filtration loss is not as dramatic for energy deposit as it is for photon flux. Knowledge of the energy deposited per laser diode pulse is important for determining fluorescence efficiencies.

The pulsed x-ray source generates a background x-ray flux in the absence of laser diode light. In the model of the light-excited x-ray tube as a single stage photomultiplier tube, this would be the x-ray flux resulting from dark current. Characterizing this background flux is necessary to determine the extent to which it contributes noise to fluorescence time spectra.

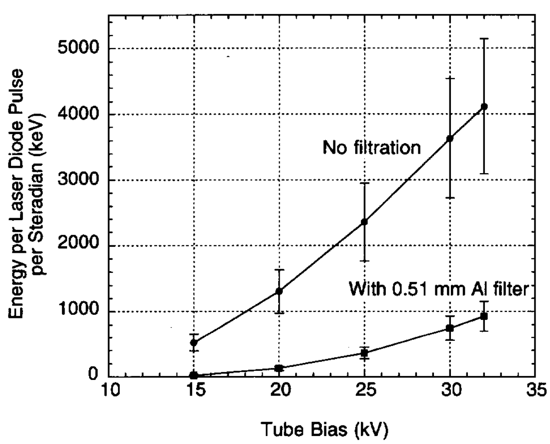

Figure 8. Mean energy deposited per laser diode pulse.

While the background flux is usually stable, a spontaneous discharge within the x-ray tube after two months of operation suddenly decreased the dark current at 30 kV bias by a factor of twenty. The dark current has subsequently increased photodiode but remains a factor of ten below the original level. Figure 9 presents pre- and post- discharge measurements of the background flux. The pre-discharge data were calculated from the current generated in a 100 µm thick photodiode placed in the x-ray beam, with a correction applied (obtained from laser-induced flux measurements) for the energy dependent detection efficiency. The post-discharge data were determined from background flux spectra with the same method used to obtain Figure 7. The background flux increases exponentially with tube bias.

Observations suggest that the background flux rate is independent of laser diode repetition rate. This has not been directly verified, but the tube cathode current at a given tube bias with the laser on appears to be the sum of the dark current at that bias (as measured with the laser off) and a value which is essentially independent of tube bias. The dark current, then, appears to be independent of laser operation, which suggests that the background flux is independent of laser operation as well.

IV. CONCLUSIONS

The pulsed x-ray source has several appealing features for the characterization of fast scintillators. It is a compact, table-top device, and is relatively inexpensive with a parts cost of about $50,000 (U.S.). It has a pulse width of less than 120 ps fwhm and with a well characterized impulse response it can be used to determine fluorescence decay times to within 50 ps. The repetition rate of the x-ray pulses can easily be varied by adjusting the laser diode pulse repetition rate. Single fluorescence photons can be detected and spectra can be averaged over time, so that accurate measurements of weak fluorescence can be made. Finally, a monochromator can readily be incorporated to select fluorescence wavelengths or to obtain fluorescence spectra.

Since the x-rays produced with this device have the same time structure as the incident light source (modulo a 31 ps fwhm broadening), this device can produce x-ray pulses with arbitrary time structure by controlling the amplitude versus time of the light source. This could be used in non-destructive testing to provide x-ray images that "freeze" the motion of moving parts within an assembly, or in medical imaging to freeze the motion of a beating heart. It could also be used to accurately characterize short (<1 ms) afterglow times for x-ray CT scanners, where an x-ray pulse is currently formed by shooting a bullet across an x-ray beam!

This pulsed x-ray source has several limitations, which include a 30 keV maximum x-ray photon energy, a low total flux, and the existence of a background flux. The 30 keV maximum x-ray energy is presently limited by electrical discharge within the x-ray tube — this could be improved significantly with alternate tube geometries. The total flux could be increased by using a more intense light source — the laser diode used here generates about 1/500 of the rated x-ray tube cathode current of 50 μA. The background flux is due to thermionic emission from the photocathode, and so can be reduced by about 2 orders of magnitude by cooling to –20° C but never eliminated. However, the background flux is quite small; the ratio of the "background" flux to the "laser on" flux is 1.5×10^{-5} with the present light source.

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Reference to a company or product name does not imply approval or recommendation by the University of California or the U.S. Department of Energy to the exclusion of others that may be suitable.

VI. References

[1] S. E. Derenzo, W. W. Moses, R. Perera, et al., "Prospects for new inorganic scintillators," *IEEE Trans Nucl Sci*, vol. NS-37, pp. 203-208, 1990.

[2] S. E. Derenzo, W. W. Moses, J. L. Cahoon, et al., "X-ray fluorescence measurements of 412 inorganic compounds," in *Conference Record of the 1991 IEEE Nuclear Science Symposium and Medical Imaging Conference*, Santa Fe, New Mexico, pp. 143-147, 1991.

[3] K. Oba, "Instrument to measure fluorescence which has occurred in a sample stimulated by x-rays," U.S. patent #4,724,536, issued to Hamamatsu Photonics, Japan, Feb 9, 1988.

[4] S.E. Derenzo, W.W. Moses, S.C. Blankespoor, M. Ito, and K. Oba, "Design of a pulsed x-ray system for fluorescent lifetime measurements with a timing accuracy of 109 ps," *Conference Record of the 1992 IEEE Nuclear Science Symposium and Medical Imaging Conference*, Orlando, Florida, Oct. 1992, pp. 117-119.

[5] S.E. Derenzo, W.W. Moses, and S.C. Blankespoor, "High Resolution Fluorescent Time Spectra of Six Ultra-Fast Powdered Inorganic Scintillators," submitted to *IEEE Transactions on Nuclear Science.*

[6] W. Yu, F. Pellegrino, M. Grant, and R.R. Alfano, "Subnanosecond fluorescence quenching of dye molecules in solution," *The Journal of Chemical Physics*, vol. 67, pp. 1766-1773, Aug. 1977.